

PTO 07-3164

PN=14-003864  
CY=JP  
DATE=20020109  
KIND=A

A TECHNIQUE AND APPARATUS FOR THE MANUFACTURE OF GAS FROM COAL AND  
SIMILAR SUBSTANCES

[石炭等を原料とする製造方法ト装置]

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Washington, D.C. March 2007

Translated by: Linguistic Systems, Inc.

PUBLICATION COUNTRY	(10): JP
DOCUMENT NUMBER	(11): 14-003864
DOCUMENT KIND	(12): A
PUBLICATION DATE	(43): 20020109
APPLICATION NUMBER	(21): 2000-187075(P2000-18705)
APPLICATION DATE	(22): 20000622
INTERNATIONAL CLASSIFICATION	(51): C10J 3/02; C10J 3/20; C10J 3/46; C10J3/48; C10K 1/14
PRIORITY COUNTRY	(33): NA
PRIORITY NUMBER	(31): NA
PRIORITY DATE	(32): NA
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TITLE	(54): A TECHNIQUE AND APPARATUS FOR THE MANUFACTURE OF GAS FROM COAL AND SIMILAR SUBSTANCES
FOREIGN TITLE	[54A]: 石炭等を原料とする製造方法ト装置

## Patent request coverage

Claim item 1: Raw materials such as coal are placed in a gasification reactor, as well as steam, water and oxygen for gas generation. The manufacture of purified hydrogen gas and the added advantage as a gas manufacturing method for carbon monoxide, as well as the provision of hydrogen, are also characteristic features of this technique.

Claim item 2: Hydrogen generated from water by electrolysis is a characteristic feature of this gas manufacturing method.

Claim item 3: Purification of part of the hydrogen and adding it to carbon monoxide and hydrogen gas, and the adjustment of the composition of the carbon monoxide and hydrogen gas are characteristic features of this gas manufacturing method.

Claim item 4: Characteristic features of this method are: The gasification reactor, which generates gas through a supply of hydrogen, oxygen from coal-like raw materials, steam and water; the electrolysis device through which water is electrolyzed from hydrogen and oxygen supplied to the gasification reactor; the purification device by which gas generated from the reactor is purified as well as carbon monoxide and hydrogen; the addition to the reactor of purified carbon monoxide gas and hydrogen gas with one part hydrogen gas; equipping of the device with a mixing vessel in which the

composition of the carbon monoxide gas and hydrogen gas is regulated.

#### Invention Details

(0001)

#### Affiliated technological fields:

The invention involves a method and device for generating hydrogen gas and carbon monoxide using oxygen and steam as well as water and hydrogen in a state of high-temperature combustion from substances such as carbides from coal and heavy oil.

(0002)

#### Present technology:

Until now, the gasification of coal and heavy oil and the creation of carbon monoxide and hydrogen was through the use of compound power fueled gas turbines with methanol ingredients. The method of generating these gases by supplying steam, water, and oxygen to coal and heavy oils results in high-temperature gases from reactions such as oxidation. Since generated gases contain carbon dioxide and sulfurated oxygen, as well as hydrogen and carbon monoxide, it is necessary to extract the hydrogen and carbon monoxide with a purification device.

A working solution: Separating gases containing carbon dioxide with a purification apparatus up to now has involved releasing it into the general atmosphere. Since carbon dioxide is one of the causes of global warming, it is highly desirable from society's point of view to reduce this. Because their generating techniques have involved reactions to oxygen and carbonized raw materials, gasification reactors up to the present time have generated a good deal of carbon dioxide. Because of this, it would be desirable to have a method that creates as little carbon dioxide as possible.

(0004)

The aim of the invention, in light of the above-mentioned problems, is to provide a coal-based gasification method and apparatus that releases the minimal amount of carbon dioxide.

(0005)

A means of resolving the problem: In order to reach the abovementioned goal, the invention in claim item 1 is a manufacturing method that uses coal-like substances in a gasification reactor as raw materials, then generates gas with steam, water and oxygen, purified hydrogen, and carbon monoxide to supply hydrogen gas.

(0006) In current gasification techniques - in which oxygen and steam was used with coal - carbon monoxide, hydrogen, and

carbon dioxide were the gases mostly generated. The chemical reactions are carried out as follows:

1.  $C + O_2 \rightarrow CO_2 + 97.7 \text{ kcal}$
2.  $C + 1/2 O_2 \rightarrow CO + 29.39 \text{ kcal}$
3.  $C + H_2 O \rightarrow CO + H_2 - 28.36 \text{ kcal}$

The heat generation reactions of Formulas 1 and 2 supply the greater part of the heat, while Formula 3 has the possibility of heat-absorbing reactions. To these formulas, the present invention adds the following reaction involving hydrogen:

- 4:  $H_2 + 1/2 O_2 \rightarrow H_2 O + 57.75 \text{ kcal}$

Formula 4 is one of heat generation reaction. When the temperature is raised in Formula 1, the reaction is suppressed, while the generation of  $CO_2$  is reduced. Formula 4 generates water vapor that can reduce the steam and water supply ratio.

(0007)

In a desirable working form, the hydrogen gas supplied will be generated by water electrolysis, through which only hydrogen gas and oxygen gas are generated, while such undesirable gases such as carbon dioxide are not. This would serve as a prevention of environmental deterioration.

(0008) Another desirable working form is through the purification of one part of the

hydrogen gas being used and adding carbon monoxide together with hydrogen, while adjusting the composition of both. With a measured volume of purified hydrogen and carbon monoxide, for example, 2:1 ( $2H_2:CO$ ), at the composition rate of methanol, it is possible to supply gas through a process similar to the generation of methanol.

(0009)

As noted in claim item 4, gas receiving supplies of both oxygen and hydrogen is generated in a gasification reactor with raw materials such as coal as well as steam and water. Hydrogen and oxygen placed the reactor are treated with a water electrolysis device, and the hydrogen and carbon monoxide generated by the reactor are purified with a purification apparatus. The purified carbon monoxide and hydrogen gas is mixed with one part of hydrogen in the reactor, and its composition is regulated in the mixing vessel.

(0010)

When materials such as coal and heavy oil, steam and water, oxygen and hydrogen are added together, as explained above and shown in the formulas, hydrogen, carbon monoxide, and carbon dioxide are generated. Aside from this, gases such as hydrogen sulphide are also generated from impurities in the mix. In the

purification apparatus, hydrogen gas and carbon monoxide are extracted, while unneeded gases like hydrogen sulphide and carbon dioxide are extracted separately. Because hydrogen and oxygen are supplied from the water in the electrolysis device, the use of an electrical power alternative such as night-time electrical power or natural energy would have fewer environmental effects, while hydrogen and oxygen could still be obtained. In the mixing vessel, part of the hydrogen supplied to the reactor is added to purified carbon monoxide and hydrogen and their composition is regulated. If the volume ratio, for example, is set at one part hydrogen and two parts carbon monoxide, it can equal the composition of methanol and supply methanol in the same capacity.

(0011)

#### Implementing the invention

An explanation for putting the device to practical use may be found by referencing the diagrams. Diagram 1 represents the gas manufacturing apparatus. The gasification reactor (1) is used for generating carbon dioxide, carbon monoxide, and hydrogen gases by adding raw materials such as coal and heavy oil with steam and water. The hydroelectrolysis apparatus (2) disassembles water with electricity, and high pressure (3-5MPa) hydrogen and oxygen generated. The gas purification device (3) purifies carbon monoxide and hydrogen, while



extracting carbon dioxide and hydrogen sulphide generated from sulfurous elements. The mixing vessel is the device in which purified hydrogen is placed and carbon monoxide and hydrogen are added in the methanol volume ratio of 1:2 in order to create the composition of methanol. Finally, the composition of coal and other raw materials for gasification, in cases in which the ratio of purified gas is not 1:2, hydrogen is supplied along with oxidized carbon in the purification apparatus (3). The methanol synthesizer (5) synthesizes methanol using the mixing vessel in which the gas is regulated. The reactivity formula of  $\text{CO} + 2\text{H}_2 \rightarrow \text{CH}_3\text{OH}$  is a heat-generating reaction with a low activity copper-based catalyst and a low temperature, and high pressure is desirable. Because of this, a high activity catalyst at less than 250 degrees centigrade and at less than 10 Mpa will synthesize methanol.

(0012)

When the device is constructed as described above, gasification reactor (1) uses raw materials such as coal and steam, with oxygen and hydrogen added as demonstrated in formulas 1 and 2, carbon dioxide and carbon monoxide are generated through heat reaction. In formula 3, carbon monoxide and hydrogen are formed. As seen in formula 4, hydrogen and oxygen react and a heat generated reaction causes the

generation of water vapor. The heat generating reaction in Formula 1 suppresses the generation of carbon dioxide. Also, because water vapor is produced, the steam supplied to the gasification reactor is reduced. Part of the generated carbon dioxide is circulated inside the reactor. Furthermore, since elements such as sulphur are also contained in the raw materials in use, hydrogen sulphide is also generated. The gasification reactor's reaction temperature as a heat-generating reaction is 850-1500 degrees Centigrade at a pressure rate of 3-5 MPa.

(0013)

Diagram 2 shows the principle behind the use of solid body macromolecule electrolyte film during electrolysis. During electrolysis, the usual decomposition method is applied by using electrolytes, but in implementing the device with simple maintenance and high purity level of the gas generated, a feasible method of creating high-pressure hydrogen and oxygen with solid body macromolecule electrolyte film is explained. Solid body macromolecule electrolyte film is distributed between an anode and a cathode, and a direct current is passed under the water. The solid body macromolecule electrolyte film works as a partition with the electrolytes, and the anode causes the reaction to occur as:  $(6) \text{H}_2\text{O} \rightarrow 2\text{H}^+ + 1/2\text{O}_2 + 2\text{e}^-$ . Hydrogen ion  $\text{H}^+$  transmits the solid body macromolecule

electrolyte film and moves it to the cathode, and the anode carries out the reaction as follows:  $(7) 2H^+ = 2e^- - H_2$ . In this way, oxygen generates from the anode and hydrogen from the cathode. The volume of oxygen generated is one-half that of hydrogen. The solid body macromolecule electrolyte film has a chemically stabilized sulfonation acid base, and a flourine resin-based cation exchange film can be used.

(0014)

Diagram 3 shows the electrolysis apparatus. In 10, the main part of how the electrolysis is carried out is shown in electrolytic cell diagram 2. In 11, in the electrolytic tank, an unadulterated tank and a tank for the separation of oxygen moisture is in position. A liquid surface is retained on the water supply. 12 is the hydrogen and water separation tank. In 13, the heat exchanger regulates the water temperature in the electrolysis tank is at a uniform degree. In 14, the non-regenerative polisher purifies the water in electrolysis tank 11. 15 is the dryer in which oxygen and hydrogen are dehydrated and separated. In 16, the relief valve for electrolysis tank 11 and for hydrogen and oxygen separation tank 12 can be opened to uniformly maintain the proper pressure strength, which is regulated at the same strength for the hydrogen and oxygen. From electrolysis cell 10 a direct electrical current flows by which the generation of hydrogen

and oxygen is commenced. When the electrical power used is an alternative source, such as night-time power or natural energy, the generation of carbon dioxide is prevented.

(0015)

In water electrolysis, if the effect of the pressure is low, the speed of reaction will be dependent on the electrical current. Because of this, even under high temperature, hydrogen and oxygen can be generated if an electrical flow is set, and it is possible without a compressor to raise the pressure to a the prescribed level. Because of this, if the pressure-endurance capacity of each tank in this device is ensured, high-pressure gas can be generated. Without the use of a compressor, putting pressure on gasification reactor (1) can produce pressurized oxygen and hydrogen.

(0016)

By using methods such as the following in the gas purification apparatus, the carbon dioxide as well as the hydrogen sulfide are removed from oxygen, with hydrogen and carbon monoxide being safely extracted:

(1) The chemical absorption process: The method using amine group attraction agents (MEA, DEA, DGA etc.), and the heated carbonic acid potassium-based attraction agent method (Benfield process, etc).

(2) The physical absorption process: Oxygen is absorbed in an organic solvent under high pressure (Selexol process, Rectisol process, etc.)

(3) The solid body adsorption process: When oxygen is at a low level of concentration, reactions such as molecular seep may be present.

(0017)

Diagram 4 shows the form of the mixing vessel. Carbon monoxide and hydrogen are placed in the vessel up to mixing gas line 20, with hydrogen added up to the hydrogen gas line 21. At each line, quantity of flow valves (24a and 24b) are provided, with gas analysis container 2(2) and quantity of flow container (23) are also provided and connected to the computer (25). From the measurement levels of gas analysis and quantity of flow containers (22 and 23) and gas lines 20 and 21, the gas composition and flow volume are calculated, and the 1:2 volume ratio of carbon monoxide and hydrogen is controlled by the computer (25), which also manages the flow volume control valves (24a and 24b). The regulated gas composition can be passed on into the methanol synthesizing chamber (5).

(0018)

In the above implementation, an explanation was given for regulating the

composition of purified hydrogen and carbon monoxide and supplying it to a methanol synthesizing device, but in the case of fueling a composite power gas turbine for this, it is possible to do this without composition regulation, and the mixing vessel is unnecessary. The method using the hydro-electrolysis device with a solid body macromolecule electrolyte film is also explained, but if there are limits to this kind of implementation, ordinary water electrolysis (the alkali hydro-electrolytic method) will suffice.

0019)

Invention efficacy: As is clear in the above explanation, the invention, by making use of coal-like substances as raw materials, with the addition of water vapor and oxygen, and results in hydrogen reactions, minimal carbon dioxide generation, and facilitates protection for the natural environment. The acquired hydrogen is generated by hydroelectrolysis, and by using night-time electrical power and natural energy as alternative power sources, it is possible to contribute to the reduction of carbon dioxide gas. Part of the hydrogen used in the gasification reactor is mixed with generated gas, and suitable uses of the composition of this gas, such as in methanol synthesizing and as composite power gas turbine fuel, are possible goals.



### Diagram Guide

Diagram 1 represents the gas generation implementation device formation.

Diagram 2 represents the hydroelectrolysis solid body macromolecule electrolyte film theory.

Diagram 3 represents the solid body macromolecule electrolyte film used in an electric device.

Diagram 4 represents the mixing vessel formation.



### Diagram Numbers Identification

- 1: Gasification reactor
- 2: Hydroelectrolysis apparatus
- 3: Gas purification apparatus
- 4: Mixing vessel
- 5: Methanol synthesizer
- 10: Electrolysis cell
- 11: Electrolysis tank
- 12: Hydrogen and water separation tank
- 13: Heat exchange chamber
- 14: Non-regenerative polisher
- 15: Dryer
- 16: Relief valve
- 20: Mixing gas line
- 21: Hydrogen gas line
- 22: Gas analysis chamber
- 23: Gas flow quantity chamber
- 24a, 24b: Flow quantity control valve
- 25: Computer







